AMENDMENT

(Amendment by Provision of the Law Article 11)

To : Examiner of the Patent Office

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1. Identification of the International Application

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4. Object of Amendment Specification and Claims

5. Details of the Amendment

(1) Specification page 3, lines 15 – 16 and 24 – 25 (English translation [0008], page 3, lines 23 – 25 and [0009], page 3/1, lines 2 – 4) "said catalyst is supported on the inner wall of a channel by covalent bond of a modified group provided on the inner wall of a channel and the

polymer" is amended as shown below.

-- said catalyst incorporated in a polymer is supported on the inner wall of a channel by covalent bond of a group provided on the inner wall of a channel or in a spacer via a group of the polymer surface --.

(2) A sentence as shown below is inserted between Specification page 4, lines 5 and 6 (English translation [0011], page 4, lines 14 – 15).

-- The surface of the inner wall of a channel preferably has silanol groups, and the spacers are covalent bonded with silanol groups by Si–O–Si bond. The groups on a polymer surface is preferably epoxide groups, and the groups in the spacers are modified with functional groups bondable with epoxide groups. --.

(3) Specification page 6, lines 27 and 29 (English translation [0022], page 7, lines 28 – 29 and 32) "amino acid group" is amended to -- amino group --. This amendment is the correction of mis-writing. Specification page 7, line 12 (English translation [0024], page 8, line 11) "amino group" is a correct writing.

(4) Claims page 13, Claim 1, lines 1 – 11 (English translation Claims page 14, Claim 1, lines 1 – 18) "A method of catalytic reaction using a micro-reactor, characterized in that:

said method of catalytic reaction uses a micro-reactor with a metal catalyst or a metal complex catalyst as a solid phase supported on the inner wall of the channel, characterized in that

said metal catalyst or said metal complex catalyst is a catalyst incorporated in a polymer,

said catalyst is supported on the inner wall of a channel by covalent bond of a modified group provided on the inner wall of a channel and the polymer,

a gas as a gas phase is passed at the center part of the channel,

a solution as a liquid phase in which a reactant is dissolved is passed between said gas and said catalyst supported on the inner wall

of a channel,

thereby the reaction of said solution and said gas is conducted by the three phase catalytic reaction of solid – liquid – gas phases accelerated by said metal catalyst or said metal complex catalyst." is amended as shown below.

-- A method of catalytic reaction using a micro-reactor, characterized in that:

said method of catalytic reaction uses a micro-reactor with a metal catalyst or a metal complex catalyst as a solid phase supported on the inner wall of the channel, characterized in that

said metal catalyst or said metal complex catalyst is a catalyst incorporated in a polymer,

said catalyst incorporated in a polymer is supported on the inner wall of said channel by covalent bond of a group provided on the inner wall of said channel or in a spacer via a group of the polymer surface,

a gas as a gas phase is passed at the center part of the channel,

a solution as a liquid phase in which a reactant is dissolved is passed between said gas and said catalyst supported on the inner wall of said channel,

thereby the reaction of said solution and said gas is conducted by the three phase catalytic reaction of solid – liquid – gas phases accelerated by said metal catalyst or said metal complex catalyst. --.

(5) Claims pages 13 – 14, Claim 8, lines 1 – 11 (English translation Claims page 15, Claim 8, lines 1 – 19) "A method of catalytic reaction using a micro-reactor, characterized in that:

said method of catalytic reaction uses a micro-reactor with a metal catalyst or a metal complex catalyst as a solid phase supported on the inner wall of the channel,

said metal catalyst or said metal complex catalyst is a catalyst incorporated in a polymer,

said catalyst is supported on the inner wall of a channel by

covalent bond of a modified group provided on the inner wall of a channel and the polymer,

hydrogen as a gas phase is passed at the center part of the channel,

a solution as a liquid phase in which a reactant is dissolved is passed between said hydrogen and said catalyst supported on the inner wall of a channel,

thereby the reaction of said solution and said hydrogen is conducted by the three phase catalytic reaction of solid – liquid – gas phases accelerated by said metal catalyst or said metal complex catalyst." is amended to

-- A method of catalytic reaction using a micro-reactor, characterized in that:

said method of catalytic reaction uses a micro-reactor with a metal catalyst or a metal complex catalyst as a solid phase supported on the inner wall of the channel, characterized in that

said metal catalyst or said metal complex catalyst is a catalyst incorporated in a polymer,

said catalyst incorporated in a polymer is supported on the inner wall of said channel by covalent bond of a group provided on the inner wall of said channel or in a spacer via a group of the polymer surface,

hydrogen as a gas phase is passed at the center part of the channel,

a solution as a liquid phase in which a reactant is dissolved is passed between said hydrogen and said catalyst supported on the inner wall of said channel,

thereby the reaction of said solution and said hydrogen is conducted by the three phase catalytic reaction of solid – liquid – gas phases accelerated by said metal catalyst or said metal complex catalyst. --.

(6) Claims page 14, after Claim 13 (English translation Claims page 16, after Claim 13) are added Claims 14 – 17 as shown below.

-- 14. (Added) The method of catalytic reaction using a

micro-reactor as set forth in claim 1, characterized in that the surface of the inner wall of said channel has silanol groups, and said spacer is covalent bonded with said silanol group by Si–O–Si bond.

15. (Added) The method of catalytic reaction using a micro-reactor as set forth in claim 1, characterized in that the group on said polymer surface is an epoxide group, and the group in said spacer is modified with a functional group bondable with an epoxide group.

16. (Added) The method of catalytic reaction using a micro-reactor as set forth in claim 8, characterized in that the surface of the inner wall of said channel has silanol groups, and said spacer is covalent bonded with said silanol group by Si–O–Si bond.

17. (Added) The method of catalytic reaction using a micro-reactor as set forth in claim 8, characterized in that the group on said polymer surface is an epoxide group, and the group in said spacer is modified with a functional group bondable with an epoxide group. --.

6. List of Papers Attached:

(1) Specification, Substitute sheets, page 3, page 3/1, page 4, page 4/1, page 6 (English translation, Substitute sheets, pages 3, 3/1, 4, 4/1, 7)

(2) Claims, Substitute sheets, page 13, 14, 14/1 (English translation, Substitute sheets, page 14, 14/1, 15, 15/1, 16)

Commun. 883 (1999)
Reference 4: M. W. Losey, and two others, Chem. Ind. Eng. Chem. Res., Vol.40, p.2555 (2001)
Reference 5: R. Akiyama and S. Kobayashi, J. Am. Chem. Soc., Vol.125, pp.3412 – 3413 (2003)
Reference 6: J. Kiji, T. Okano, Y. Higashimae, and Y. Fukui, Bull. Chem. Soc. Jpan., Vol.69, pp.1029 – 1031 (1996)

However, such three phase catalytic reactions as three phase catalytic reductive reactions of solid – liquid – gas phases using a heterogeneous catalyst have never so far been effectively realized by a micro-reactor.

Disclosure of the Invention

[0008] In view of the problems mentioned above, it is an object of the present invention to provide a method of catalytic reaction using a micro-reactor capable of conducting three phase catalytic reaction of solid – liquid – gas phases in short time at high yield.

In order to attain the above-mentioned object, the present invention is a method of catalytic reaction using a micro-reactor with a metal catalyst or a metal complex catalyst supported as a solid phase on the inner wall of a channel, characterized in that the metal catalyst or the metal complex catalyst is a catalyst incorporated in a polymer, said catalyst incorporated in a polymer is supported on the inner wall of a channel by covalent bond of a group provided on the inner wall of a channel or in a spacer via a group of the polymer surface, a gas as a gas phase is passed at the center part of the channel, a solution as a liquid phase in which a reactant is dissolved is passed between the gas and the catalyst supported on the inner wall of a channel, thereby the reaction of the solution and the gas is conducted by the three phase catalytic reaction of solid – liquid – gas phases accelerated by the metal catalyst or the metal complex catalyst. In the above-described aspect, the gas phase preferably consists of hydrogen or carbon monoxide.

[0009] Also, another aspect of the present invention is a method of

catalytic reductive reaction using a micro-reactor with a metal or a metal complex catalyst supported as a solid phase on the inner wall of a channel, characterized in that the metal catalyst or the metal complex catalyst is a catalyst incorporated in a polymer, said catalyst incorporated in a polymer is supported on the inner wall of a channel by covalent bond of a group provided on the inner wall of a channel or in a spacer via a group of the polymer surface, hydrogen as a gas phase is passed at the center part of the channel, a solution as a liquid phase in which a reactant is dissolved is passed between hydrogen and the catalyst supported on the inner wall of a channel, thereby the reaction of the solution and hydrogen is

conducted by the three phase catalytic reaction of solid – liquid – gas phases accelerated by the metal catalyst or the metal complex catalyst. According to the above-described aspect, hydrogenation reaction, hydrocracking reaction, or carbon monoxide insertion reaction of various substances can be conducted by three phase catalytic reaction in short time at high yield.

[0010] In the above-described aspect, the metal catalyst is preferably palladium. The metal complex catalyst is preferably a palladium complex catalyst.

[0011] The metal catalyst may be either chromium, manganese, iron, cobalt, nickel, copper, molybdenum, ruthenium, rhodium, tungsten, osmium, iridium, or platinum. The metal complex catalyst may be a metal complex catalyst of either chromium, manganese, iron, cobalt, nickel, copper, molybdenum, ruthenium, rhodium, tungsten, osmium, iridium, or platinum. The surface of the inner wall of a channel preferably has silanol groups, and the spacers are covalent bonded with silanol groups by Si–O–Si bond. The groups on a polymer surface is preferably epoxide groups, and the groups in the spacers are modified with functional groups bondable with epoxide groups.

[0012] According to the present invention, three phase catalytic reductive reaction can be conducted in short time by supporting a catalyst, particularly a metal or a metal complex catalyst as a solid phase on the inner wall of a micro-channel of a micro-reactor. Further, since such complicated operation as separation of products and a catalyst and recovery of a catalyst is unnecessary, continuous operation of long time is possible.

Brief Description of the Drawings

[0013]

Fig.1 diagrammatically illustrates the makeup of a micro-reactor used in the embodiments of the present invention, and (a) is a plan view and (b) is a cross-sectional view along a line Y – Y.

Fig. 2 is a cross-sectional view illustrating the states of a solution and hydrogen passing through a micro-channel of the micro-reactor used in the present invention.

Fig. 3 is a view diagrammatically illustrating the reaction to

support a PI catalyst in a micro-channel.

its inner wall 4c. The reaction mixture containing the object formed by reaction is collected into a vessel for recovery 10, and taken out upon necessity.

[0020] Here in the solid – liquid – gas phase reaction, hydrogenation of the reactant, that is, catalytic reductive reaction in case of hydrogen as the gas phase, and such catalytic reactions as carbon monoxide insertion reaction into the reactant, for example, carbonylation reaction in case of carbon monoxide as the gas phase can be caused.

[0021] As the solid catalyst 5 used for the solid – liquid – gas phase reaction, a metal or a metal complex catalyst of either of palladium (Pd), Chromium (Cr), manganese (Mn), iron (Fe), cobalt (Co), nickel (Ni), copper (Cu), molybdenum (Mo), ruthenium (Ru), rhodium (Rh), tungsten (W), osmium (Os), iridium (Ir), or platinum (Pt) may be used.

[0022] The above-mentioned catalyst 5 is preferably a polymer-inclusion catalyst (hereinafter to be called a PI catalyst) with the above-mentioned metal catalyst or metal complex catalyst immobilized in a polymer (Refer to the above-mentioned Reference 5.). In order to firmly fix the PI catalyst 5 not to dissociate from the inner wall 4c of the micro-channel, it is preferred to immobilize, that is, to support by a covalent bond. For that, in case that the inner wall 4c of the micro-channel is glass, one end of a spacer 4d of the PI catalyst 5 mentioned below is modified with a trialcoxysilane structure, and bonded to a silanol group on the glass surface as the inner wall 4c of the micro-channel. The other end of the spacer 4d can be bonded directly to, for example, an epoxy group on the polymer surface of the PI catalyst 5 by modifying it with a functional group such as amino group and others. In case that the inner wall 4c of the micro-channel is a resin, bonding is similarly possible to the above-mentioned epoxy group by modifying the resin surface with a functional group such as amino group and others.

Since thereby the PI catalyst can be firmly supported on the inner wall 4c of a micro-channel, it does not dissociate from the inner wall 4c of a micro-channel, and can be used repeatedly.